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Charge transport in MDMO-PPV:PCNEPV all-polymer solar cells

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Charge transport properties are investigated of blends of poly [2-methoxy-5-(3′, 7′-dimethyloctyloxy)-1,4-phenylene vinylene] (MDMO-PPV) and poly-[oxa-1,4-phenylene-(1-cyano-1,2-vinylene)-(2-methoxy-5-(3′, 7′-dimethyloctyloxy)-1,4-phenylene)-1,2-(2-cyanovinylene)-1,4-phenylene] (PCNEPV). The hole transport in the MDMO-PPV donor phase of the 1:1 weight ratio blend is trap-free space-charge limited, with a mobility identical to the pristine polymer. The electron current in the PCNEPV acceptor phase is strongly reduced by traps that are exponentially distributed in energy. The current in MDMO-PPV:PCNEPV bulk heterojunction solar cells is therefore unbalanced and dominated by the holes in the MDMO-PPV phase. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2734101]

I. INTRODUCTION

Polymer blends are potential candidates for solar energy conversion, due to their flexibility, ease of processing, and low costs. Presently, the most efficient organic solar cells are made of polymer:fullerene blends, with efficiency values from 3.5 to more than 4%.[1–4] In these bulk heterojunctions (BHJ) excitons are created in the polymer phase, which dissociate at the interface between the polymer donor phase and fullerene acceptor phase. The resulting geminate pair at the interface consists of a hole in the donor and an electron in the acceptor, which needs to be separated into free-charge carriers with the help of an electric field. Subsequently, the free holes and electrons are transported through the donor and acceptor phase of the blend, respectively, toward the electrodes, giving rise to an external photocurrent. During the charge transport process, free electrons and holes might recombine again, leading to losses in the performance. Consequently, charge transport is a crucial process in polymer BHJ solar cells. A remarkable feature of BHJ solar cells based on poly [2-methoxy-5-(3′, 7′-dimethyloctyloxy)-1,4-phenylene vinylene] (MDMO-PPV) and [6,6]-phenyl C_{61}-butyric acid methyl ester (PCBM) is that the optimum performance of 2.5% is reached adding up to 80 wt % of PCBM, a hardly absorbing material in the solar spectral region.[5] The necessity of such a large amount of PCBM arises from a strong enhancement of the hole transport in MDMO-PPV when blended with PCBM,[6] combined with an enhanced dissociation efficiency due to the increase of the dielectric constant.[7] For PPV-based compounds with lower hole mobilities, it was demonstrated that the photocurrent reaches a fundamental space-charge limit, which is detrimental for the fill factor and efficiency.[8]

An attractive alternative for the relatively poor absorption of the fullerenes is the use of conjugated polymers as the electron accepting unit. In these all-polymer blends, light is absorbed in both the donor and the acceptor phase.[9] Initially, solar cells made with blends of poly (dialkoxy-*p*-phenylene vinylene) (PPV) derivatives have given efficiencies lower than 1%.[10,11] In recent studies of blends of PPV-based donors together with a cyano (CN) substituted PPV[12] or a red-emitting polyfluorene[13] as acceptor, efficiencies exceeding 1.5% have been reported, thus approaching the efficiency of their fullerene counterparts.[5] Regarding the device operation of all-polymer cells, it has been shown by Veenstra *et al.* that blends of poly [2-methoxy-5-(3′, 7′-dimethyloctyloxy)-1, 4-phenylene vinylene] (MDMO-PPV) and poly-[oxa-1, 4-phenylene - (1-cyano-1,2-vinylene) - (2-methoxy-5-(3′, 7′-dimethyloctyloxy)-1, 4-phenylene)-1, 2-(2-cyanovinylene)- 1, 4-phenylene] (PCNEPV) form an efficient charge-transfer donor-acceptor system, due to the difference between the electron affinities (~0.5 eV) and the ionization potentials (~0.7 eV) of the two polymers.[11] The solar cells made of

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this blend give a large open circuit voltage (V_{oc}) of about 1.3–1.5 V. However, the optimized device shows an efficiency limited to about 0.75%, possibly due to a reduced charge transport.[11] So far, information about the charge transport properties of all-polymer BHJ solar cells is missing. In the present study we report on the electron and hole transport in blends consisting of MDMO-PPV as donor and PCNEPV as acceptor. We observe that the hole mobility in the PPV phase of the blend, in contrast to MDMO-PPV:fullerene blends, is not enhanced upon blending with PCNEPV. Furthermore, the electron transport in the PCNEPV phase is strongly reduced by traps, leading to an unbalanced charge transport.

II. EXPERIMENTAL PROCEDURE

A typical all-polymer bulk heterojunction (BHJ) solar cell is a multilayer structure, with the polymer blend sandwiched between two electrodes. Poly (3,4-ethylenedioxythiophene):poly(4-styrene sulphonate) (PEDOT: PSS), which matches the highest occupied molecular orbital (HOMO) of the electron donor MDMO-PPV, is spin-coated on transparent indium tin oxide (ITO) coated glass to create an ohmic contact for holes. For electrons, LiF/Al forms an ohmic contact with the lowest unoccupied molecular orbital (LUMO) of the electron acceptor (PCNEPV). However, in order to distinguish between the electron and hole transport, test devices are necessary that either block the hole or electron injection into the blend. For hole transport studies the electron current in the blend is suppressed by the use of a palladium (Pd) electron blocking top contact.[7] For electron transport studies in the blend, the suppression of holes injection is established by using a low work function, hole-blocking bottom electrode. However, low work function metals such as Ba and Ca are often reactive and incompatible with processing from solution. Alternatively, a nonreactive and low work function electrode can be obtained by modifying the silver (Ag) work function with a self-assembled monolayer of hexadecanethiol. In this way, the silver work function is lowered from 4.3 to ~3.8 eV, creating a large hole injection barrier of ~1.5 eV with respect to the HOMO of MDMO-PPV. This enables the measurement of the electron-only current in the blend.[14]

The electron donor, MDMO-PPV, was synthesized using the sulfinyl route,[15] yielding a molecular weight of 300 kg/mol and a polydispersity index of 2.7.

As electron acceptor, PCNEPV was synthesized as described elsewhere,[16] with a molecular weight of 73.4 kg/mol and a polydispersity index of 3.3. The molecular weights and polydispersity indexes were measured against poly(styrene) standards.

The double-carrier and hole-only devices were prepared on ITO coated glass substrates, provided by Philips Research. For electron-only diodes, glass slides were used. All substrates were cleaned, dried, and treated with UV-ozone prior to PEDOT:PSS (Bayer AG) spin-coating or Ag deposition. After spin-coating, the PEDOT:PSS layer was dried at 140 °C for 10 min, and subsequently, the polymer blend (1:1 weight ratio) was spin-coated from chlorobenzene solution in a nitrogen atmosphere. For hole-only diodes a palladium top electrode was deposited at ~10^{-6} mbar, with a thickness of about 20 nm, covered by a gold layer of 60–80 nm.

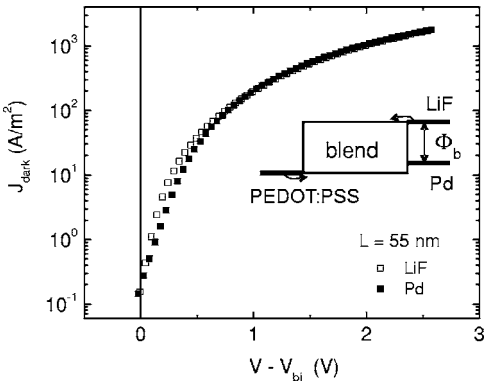

FIG. 1. Dark *J-V* characteristics of a solar cell (LiF/Al electrode, open symbols) and of a hole-only blend device (Pd electrode, filled symbols). For both devices, the active layer is 55 nm thick, and the *J-V* characteristics have been corrected for the corresponding built-in voltage. The inset shows the schematic energy band diagram of the devices.

The electron-only diodes were prepared on glass substrates, on which Ag was deposited after the UV-ozone treatment. For a better adhesion, 1 nm of chromium (Cr) was first deposited on glass by thermal evaporation, followed by 50 nm of Ag, at a chamber pressure of ~10^{-6} mbar. The substrates were then immersed in a hexadecanethiol ($HSC_{16}H_{33}$) solution of ~3×10^{-3} M in ethanol and kept for at least 36 h, then rinsed with ethanol, toluene and *iso*-propanol, and dried. Subsequently, the polymer blend was spin-coated on the substrates, under a nitrogen atmosphere. As a top electrode, 5 nm barium (Ba) or 1 nm LiF, deposited at a chamber pressure of ~10^{-6}–10^{-7} mbar, followed by 100 nm of Al, was used for the electron-only and double-carrier devices.

The devices were characterized using a Keithley 2400 SourceMeter, the measurements being performed under nitrogen atmosphere. The light output of the double-carrier devices have been recorded with a photodiode connected to a Keithley 6514 electrometer. The bottom electrode work function has been measured with a Kelvin probe.

III. RESULTS AND DISCUSSION

A. Hole transport in the blend

A direct indication of the dominance in transport of one type of carrier in the blend is obtained by comparing a double-carrier and a hole-only device. In Fig. 1 the *J-V* characteristics are shown for a PEDOT:PSS/MDMO-PPV:PCNEPV (1:1)/Pd hole-only and a PEDOT:PSS/blend/LiF/Al double-carrier device, both with a thickness of 55 nm. It appears that the current of the double-carrier device is completely hole dominated, as the current coincides with the one of the hole-only device. The presence of electrons in the device does not significantly change the magnitude of the charge transport.

Furthermore, the hole mobility in the blend is compared with the mobility of the pristine polymer. In the case of MDMO-PPV:PCBM blends, the hole mobility of the MDMO-PPV phase increases about 400 times as compared

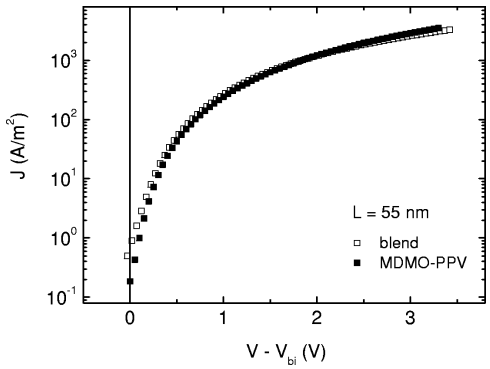

FIG. 2. *J-V* characteristics (dark) of a blend hole-only device (empty symbols) and of pristine MDMO-PPV hole-only device (filled squares). In both cases, the active layer thickness is 55 nm.

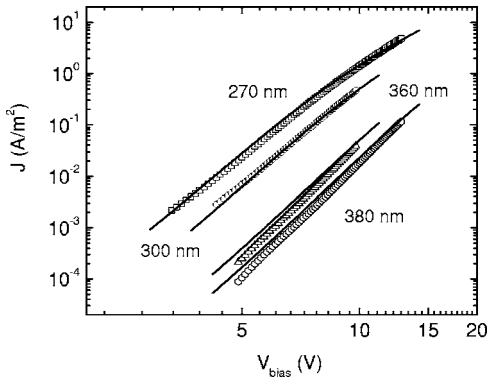

FIG. 3. Thickness-dependent electron *J-V* characteristics of PCNEPV in the blend for active layers of 270 nm (squares), 300 nm (circles), 360 nm (triangles), and 380 nm (hexagon) and the calculated lines using the exponential trap model with $N_t=9.6\times10^{22}$ m^{-3} and $T_t=2500$ K.

with the pristine polymer when blended with 80 wt % PCBM. This enhancement is also responsible for the relatively high efficiencies of the system.[6] However, as shown in Fig. 2, the hole current of the MDMO-PPV:PCNEPV blend is equal to the hole current of a pristine PEDOT:PSS/MDMO-PPV/Pd hole-only device. As a result, the hole transport in the blend is similar to that in the pristine MDMO-PPV.

The hole transport in pristine MDMO-PPV is known to be space-charge limited, enabling a direct determination of the mobility. The hole-only current in the MDMO-PPV is described by a space-charge limited current with a zero-field mobility at room temperature (RT) of 5×10^{-10} m^2/V s. Analyzing the temperature dependence of the MDMO-PPV hole mobility within the Gaussian disorder model[17] gives a width of the Gaussian density of states (DOS) of 0.109 eV. In the modeling, a relative dielectric constant of 2.1 has been assumed for MDMO-PPV.[18]

B. Electron transport in the blend

The electron current in the PCNEPV phase of the blend is measured in an electron-only diode using a nonreactive Ag/SAM bottom electrode and an ohmic top contact as LiF/Al or Ba/Al. In MDMO-PPV the electron-only current exhibits a strong dependence on applied voltage as well as sample thickness, which is a fingerprint for trap-limited transport with an exponential energy distribution of traps.[19] For a field- and density-independent mobility, the current density is given by[20]

$$J=N_c q\mu_n\left(\frac{\varepsilon_0\varepsilon_r}{qN_t}\right)^r\left[\left(\frac{2r+1}{r+1}\right)^{r+1}\left(\frac{r}{r+1}\right)^r\right]\frac{V^{r+1}}{L^{2r+1}}, \qquad (1)$$

with q the elementary charge, $\varepsilon_0\varepsilon_r$ the dielectric constant, V the applied voltage, L the sample thickness, μ_n the free-electron mobility, N_c the effective density of states, N_t the amount of traps, and $r=T_t/T$, where T_t is the characteristic temperature of the exponential distribution.

In Fig. 3 the electron current in the PCNEPV phase is shown for a variety of thicknesses. Identical to MDMO-PPV, also the electron current in the PCNEPV exhibits a strong dependence on voltage and sample thickness. We note that the electron current in PCNEPV phase is about one order of magnitude higher than the electron current in MDMO-PPV, ensuring that the measured electron current is dominated by the PCNEPV.

As a final step we use the free-carrier mobility $\mu_n(n,E,T)$ determined for PCNEPV, as discussed in the next section, to describe the trap-limited electron current in MDMO-PPV:PCNEPV blends shown in Fig. 3. The measured electron currents are modeled for different thicknesses assuming an exponential distribution of traps, with $N_t=9.6\times10^{22}$ m^{-3} and $T_t=2500$ K. With $\mu_n(n,E,T)$, N_t, and T_t known, also the temperature dependence of the trap-limited current electron is completely fixed. In Fig. 4 it is demonstrated that this set of parameters also consistently describes the temperature dependence of the electron current in the PCNEPV phase in the blend.

C. Mobility of free carriers in PCNEPV

In order to model the electron current of the PCNEPV, we cannot directly apply Eq. (1) to the experimental *J-V* characteristics of Fig. 3. First, the prefactor contains both the mobility of the free carriers as well as the amount of traps. These two parameters cannot be disentangled from the electron-only *J-V* characteristics. Second, in conjugated polymers the mobility is known to depend on the charge carrier density and electric field,[21,22] whereas Eq. (1) is only

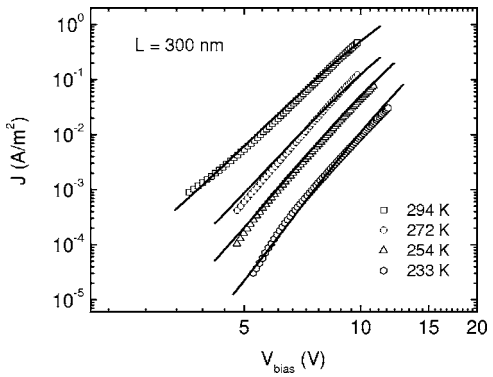

FIG. 4. Temperature-dependent electron *J-V* characteristics (symbols) and the calculations (lines) using the exponential trap model with $N_t=9.6\times10^{22}$ m^{-3} and $T_t=2500$ K. The active layer thickness of the device is 300 nm.

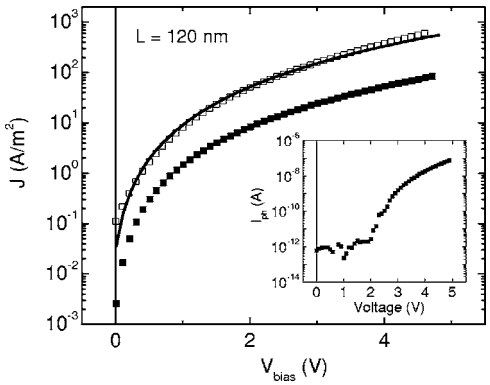

FIG. 5. Experimental LED current density vs voltage characteristics at RT (empty symbols) of PCNEPV, the calculated double-carrier current with a density-dependent free-carrier mobility (line), and the calculated hole-only current (filled symbols) for the same mobility value. The zero-field mobility value assumed in the simulation is 6×10^{-11} m^2/V s, and for the double-carrier current the electron trapping also has been included. The inset shows the measured light output (photocurrent) of the LED.

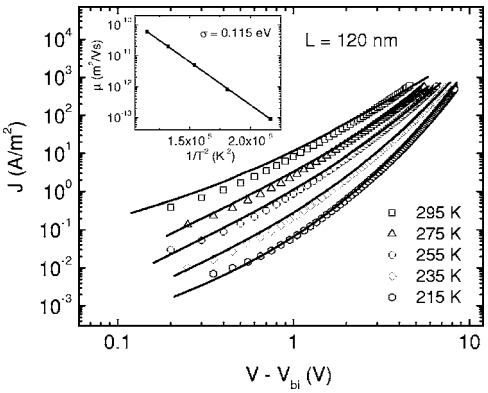

FIG. 6. Temperature-dependent *J-V* characteristic of a PCNEPV double-carrier (LED) device (symbols) and the calculations (lines) with a free-carrier density-dependent mobility and electron trapping. At low temperatures, the field dependence of mobility has been included. The inset shows the temperature-dependent zero-field mobility values obtained from the simulation (symbols) and the calculated line using the correlated Gaussian model (CGM), with $\sigma_{DOS}=0.115$ eV.

valid for a constant mobility. However, from the observed dependence of the electron current on voltage and sample thickness we can already estimate that T_t is about 2500 K. For MDMO-PPV the dependencies of the hole mobility $\mu_p(p,E,T)$ on carrier density (p), electric field (E), and temperature (T) are known from measurements on hole-only diodes.[21] Furthermore, it has been shown by Chua *et al.* that in transistors electrons and holes have similar mobilities.[23] By assuming that $\mu_n(n,E,T)$ is equal to $\mu_p(p,E,T)$, the electron currents in MDMO-PPV can be numerically modeled.[19] For PCNEPV there are no studies on the hole transport available; a major problem is that the HOMO of PCNEPV is about 6 eV below the vacuum level. As a result, the hole currents injected from standard anodes as Au (~5 eV) or PEDOT:PSS (~5.1 eV) are not space-charge limited but strongly injection limited, so a direct determination of the mobility is not possible. In order to obtain information about the hole transport in PCNEPV, we make use of an effect that has been observed in polyfluorene that also has its HOMO level located at about 6 eV. In a double-carrier PEDOT:PSS/PFO/Ba/Al light-emitting diode, the presence of electrons electrically tailors the PEDOT:PSS/PFO contact, transforming it into an ohmic contact.[24,25] The idea is that the segregation of the PSS toward the PEDOT:PSS surface creates a thin insulating barrier of polystyrene sulphonic acid (PSSH) at the polymer interface, and also a deep electron trap. The electrons injected in the polymer are trapped at this interface, charging the thin insulating barrier and therefore enhancing the tunneling of holes into the polymer.

We use the same approach to determine the mobility of free carriers in a double-carrier LED of PCNEPV. After electrical conditioning of the interface between PEDOT:PSS and PCNEPV light emission occurs (inset of Fig. 5) already after crossing the LED built-in voltage of around 2 V, as expected from the PCNEPV band gap and the presence of two ohmic contacts. The quadratic dependence of current density on voltage at RT is a further sign of trap-free space-charge limited behavior, in which the holes are the dominant carriers (as the electron current gives much stronger voltage dependence).

Under the assumption that $\mu_n(n,E,T)=\mu_p(p,E,T)$, we can model the double-carrier current and unambiguously extract the mobility, as shown in Fig. 5. It should be noted that the double-carrier current is nearly independent on the amount of electron traps.[19] From the quadratic part of the *J-V* characteristics, a zero-field mobility at RT of 6×10^{-11} m^2/V s is directly obtained. The high-voltage part at room temperature can be described using a density-dependent mobility of the form[21]

$$\mu_h(p,T)=\mu_h(0,T)+\frac{\sigma_0}{e}\left[\frac{\left(\frac{T_0}{T}\right)^4\sin\left(\pi\frac{T}{T_0}\right)}{(2\alpha)^3B_c}\right]^{T_0/T}p^{(T_0/T)-1}, \tag{2}$$

with the following parameters: a conductivity prefactor $\sigma_0=7.5\times10^{-7}$ S/m, an effective overlap parameter between localized states $\alpha^{-1}=0.14$ nm, a width of the exponential density of states $T_0=600$ K, and the critical number for the onset of percolation $B_c=2.8$. Furthermore, from impedance measurements, the relative dielectric constant of PCNEPV has been calculated to be 3.2.

We also evaluated the temperature dependence of the LED current, shown in Fig. 6. From the zero-field mobility, obtained from the low-voltage part, a width of the Gaussian DOS for PCNEPV has been determined to be 0.115 eV by applying the correlated Gaussian disorder model.[26] At low temperatures also a field dependence of the mobility has been taken into account.[21,22]

D. DISCUSSION

With the electron and hole transport in the MDMO-PPV:PCNEPV blends characterized, we now can make a direct comparison with the transport in the commonly used MDMO-PPV:PCBM (1:4 wt %) blend. For MDMO-PPV:PCBM, values of $\mu_e=2.0\times10^{-7}$ m^2/Vs and $\mu_h=1.4\times10^{-8}$ m^2/Vs have been found,[7] which are considerably larger than the values found in the MDMO-PPV:PCNEPV blends. Regarding holes, the current in the all-polymer blend

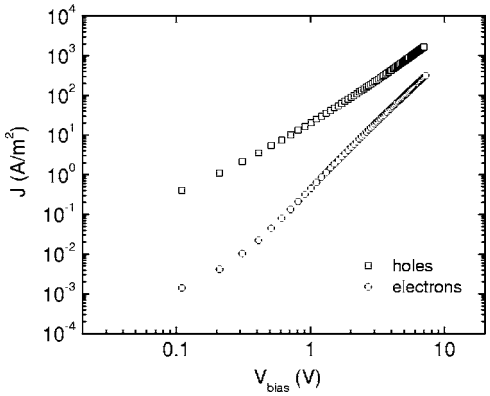

FIG. 7. Calculated *J-V* characteristics of a hole-only device (squares), and of an electron-only device (circles), for an active layer thickness of 100 nm. The values of the parameters used to calculate the *J-V*s are the one derived from charge transport measurements.

is not enhanced by two orders of magnitude, as in the polymer-fullerene blend. The electron mobility of 6×10^{-11} m^2/V s in the PCNEPV is not only more than three orders of magnitude lower than the electron mobility in PCBM, but, moreover, the transport is further hindered by the presence of traps. In Fig. 7, the hole and electron transport in the MDMO-PPV:PCNEPV blend are compared, showing that the presence of traps leads to a strongly unbalanced transport. For the solar cells it is clear that the extraction of charge carriers will be strongly hindered by the low mobilities and the presence of traps. This reduced extraction is expected to lead to increased recombination losses. Furthermore, the difference in electron and hole transport of about two orders of magnitude at low voltages (<1 V) will lead to the formation of space charges. A detailed evaluation of the photocurrents generated in these blends is a subject for further study.

IV. CONCLUSION

We have investigated the charge transport in blends of MDMO-PPV as donor and PCNEPV as acceptor. The hole transport in the MDMO-PPV phase is space-charge limited and the hole mobility is equal to the value of the pristine polymer. The electron transport in the PCNEPV phase is strongly trap-limited. The presence of electron traps leads to a highly unbalanced charge transport in this type of blend.

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